LCA Case Studies

Recycling of EOL CRT Glass into Ceramic Glaze Formulations and Its Environmental Impact by LCA Approach

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Abstract

Background, Aims and Scope. The interest in recycling materials at the end of their life is growing in the industry in general. As regards the Wastes of Electrical and Electronic Equipment (WEEE), an appreciable increase of these materials has been noticed in the last decades, $117 \cdot 10^3$ tons of WEEE have been produced in Italy in 2002 according to Ecohitech [1] and the increase in this kind of waste is three times higher than that of the municipal waste according to the FISE ASSOAMBIENTE report [2]. Within WEEE, End-of-Life Cathode Ray Tube (EOL CRT) glass, the main part of TV sets and PC monitors, is here analysed using both a technical approach to establish a possible reuse of the glass in a open-loop recycling field (ceramic industry) and a methodology (LCA) capable of providing environmental evaluations.

Methods. The technological characterization was performed by chemical resistance tests (UNI EN ISO 10545-13), staining tests (UNI EN ISO 10545-14) with blue methylene and potassium permanganate ($KMnO_4$), and surface abrasion tests (UNI EN ISO 10545-7). The LCA study was conducted using the SimaPro 5.0 software and Eco-Indicator 99 as an evaluation method.

Results and Discussion. The good technical results, reached by using cleaned EOL CRT panel glass inside a ceramic glaze formulation instead of a commercial frit, are supported by the environmental impact evaluation, which shows a decrease of the overall potential damage (measured in Points) of 36% and, in particular, a reduction of 53% in 'Human health', 31% in 'Ecosystem quality' and 24% in 'Resources'.

Conclusions. This study has demonstrated that this new, open-loop recycling strategy for the CRT glass significantly reduces the environmental impact of the ceramic glaze production process. In fact, in all damage categories examined in this study, there is a minor impact. An improvement is evident in the respiratory inorganics sub-category related to the lowering of dusts mainly and to a lesser amount with NO_x and SO_x in the climate change sub-category, due mainly to the reduction of CO_2 emission correlated to the avoided combustion of the mixture which feeds melting furnaces in the frit production. Thus, the damage decrease in 'Ecosystem quality' is prevalently due to the lower NO_x emissions by the kilns in the frit production that is evident in the acidification/eutrophication sub-category. Finally, the significant saving in the 'Resource' category is principally linked to the fossil fuels sub-category, thanks to the methane saving which stokes the melting furnaces.

Perspectives. Furthermore, the decrease in CO_2 emission (94.4%) evident in the climate change sub-category is a very important topic because it is in line with the Kyoto protocol (1997), where significant efforts have been exerted for the reduction of the green house gases emission, notably CO_2. The CO_2 emission is correlated to the combustion of the mixture which feeds melting kilns in the frit production, therefore the recycling of secondary raw materials, already in a glass state, can reduce the emissions of this gas. This reduction can be termed as environmental credit and it is an example of an allocation of environmental loads in a open-loop recycling, where waste from one industrial system are used as raw materials in another product system.

Keywords: Cathode ray-tube glass; ceramic glaze; life cycle assessment; recycling

1 Waste of Electrical and Electronic Equipment (WEEE) Problematics

The waste stream of electrical and electronic equipment has been identified as one of the fastest growing waste streams in the European Union. Today, this stream, calculated as 7.5 millions tons in the EU [3], constitutes 4% of the municipal waste, and is assumed to increase by 16–28% every five years. This represents a growth three times faster than the average municipal waste [4]. In order to reduce the amount (around 90%) of electrical and electronic waste disposed of in landfills incinerated or recovered without any preventive treatment, the waste classification and the WEEE directive seek to establish separate collection and recycling systems for electrical and electronic waste. The recent Decision of the European Communities 2000/532/EC classifies discarded electrical and electronic equipment containing hazardous components, such as glass from cathode ray tubes, as hazardous waste which cannot be landfilled. Further, the WEEE directive implements the principle for producers to take into account, already at the product design stage, as well as the need to reduce the use of hazardous substances (2002/95/CE) and to improve the recyclability of electrical products (2002/96/CE). These decisions will increase challenges to find out possible reuses for End-of-Life (EOL) Cathode Ray Tube (CRT) glass, the main part of TV sets and PC monitors.

2 The Glass Components of a Cathode Ray Tube (CRT)

The CRT or kinescope, which is composed of 85% glass and 15% plastic and metal, represents about two thirds of the weight of a television set or a computer monitor. The recycling techniques for EOL CRT glass is quite problematical because of the different components which constitute themselves (**Fig. 1**):

1. Panel (the front part): a barium, strontium and very homogeneous glass, of a greenish-blue colour, whose weight is about 2/3 of the whole CRT
2. Funnel (the hidden part inside the TV set): a lead glass, whose weight is about 1/3 of the whole CRT
3. Frit (the connection between the panel and the cone): a low melting temperature lead glaze
4. Neck: a very rich lead glass that envelops the electron gun.

However, it is important to point out that various different compositions of glasses used for CRT are available. These mainly depend on: the Country manufacturing the product, the period in which the CRTs were produced, and whether the CRT is to be used for a colour or black & white TV set or a PC monitor. Because of these factors, it would be more correct to consider a range of compositions such as the one shown in **Table 1** [5].

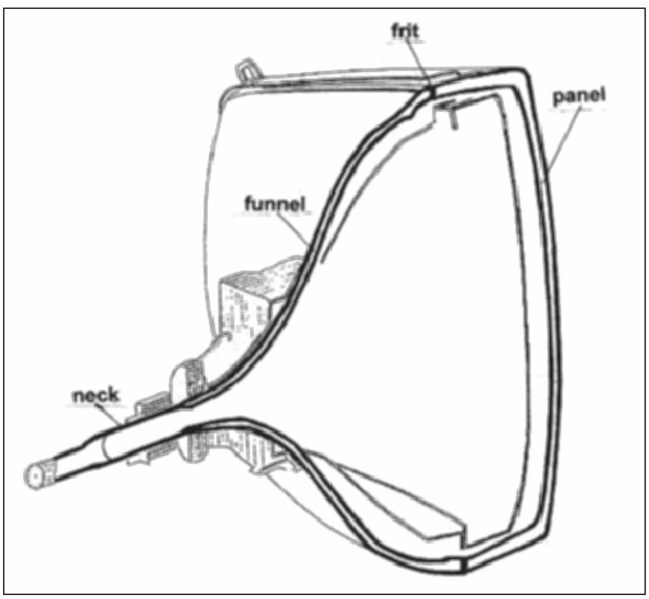

Fig. 1: Cathode Ray Tube (CRT) for TV set or PC monitor [2]

Table 1: Chemical composition ranges of panel and funnel glass for CRTs

wt%	Panel [a]	Funnel
SiO_2	59.0–64.5	50.30–63.80
Al_2O_3	1.75–3.65	1.1–5.0
Na_2O	5.15–9.45	5.3–8.3
K_2O	6.0–8.5	6.1–10.3
CaO	0.05–4.35	1.05–4.45
MgO	0.1–1.75	0.5–3.0
BaO	2.2–13.9	0.05–3.7
SrO	0.2–11.6	0.08–1.0
PbO	0.0–2.8	11.0–23.6
ZrO_2	0.0–3.5	0.0–0.5

[a] Traces of (<1%) of LiO_2, ZnO, As_2O_3, Ti O_2, F and CeO_2

For all the above-mentioned problematics, if CRT glasses are crushed and mixed together, they cannot be recycled as cullet, for instance for industrial glass production such as container glass, tableware glass, TV glass, etc. On the other hand, if the different components are separated and cleaned from the coating by applying suitable treatments belonging to some specialised industries, it is possible to obtain a secondary raw material of great quality, usable both in the CRT glass-making process (closed-loop recycling) and in other applications (open-loop recycling).

In developing closed-loop recycling, some CRT producers have made remarkable progress: at the moment more than 35% of EOL CRT glass can be used in the funnel glass and at least 15% in the panel one [6]. Higher recycling rates are being tested at the moment, but closed-loop recycling cannot still utilise all the material coming from the market, also considering the coming of the digital technology that will reduce this use. As regards open-loop recycling, some difficulties are related to the prohibition to introduce dangerous elements (such as lead) into products like glass containers, domestic glassware or glass fibres. So, the glass industry is an excellent potential user of carefully selected glass for screens and therefore without the above-mentioned elements. In the ceramic industry, instead, the limitation is not so restrictive and both glass from screens (panel) and glass from cones (funnel) are potentially usable, even if they must be supplied with particular characteristics of homogeneity, cleanness, etc. In particular in this paper panel glass was only considered for the obtainment of a final product. This choice is related to the presence of panels in large quantities (65% of the weight of a CRT), to its great versatility linked to the lead absence and to its utilisation in low quantities (15%) in closed-loop recycling.

3 CRT Glass as a Glaze Raw Material

A glaze is defined as a continuous adherent layer of glass (or glass and crystals) on the surface of a ceramic body that is hard, non absorbent, and easily cleaned. The surface may be shiny or matte. A glaze is usually applied as a suspension of glaze-forming ingredients in water. After the glaze layer dries on the surface of the piece, it is fired, whereupon the ingredients melt to form a thin layer of glass. The glaze may be fired at the same time as the body or in a second firing. Glaze suspensions are composed of different types of raw materials, which can be divided into three groups: non plastics, plastics and additives. The non-plastic materials include oxides, pigments, feldspars and others principal components such as frits. The frits are obtained from mixtures of silicates, carbonates and oxides which are melted and rapidly cooled in water. Plastic materials consist of clays, generally kaolin and bentonite. Finally, the additives used to optimise the glazing process are dispersing and deflocculanting agents, binders and defoaming agents [7].

By looking at the CRT glass composition (in particular that of panel glass), it appears evident that the purified CRT glass is a very valuable raw material, especially in applications where homogeneous silicate compositions are desired. If starting from crystalline raw materials like sand and alkali carbonates (for instance to melt a glassy frit), the formation and mixing of silicates to a homogeneous material is a very energy-consuming process. Thus, EOL CRT cleaned glass could represent an important raw material able to help to reduce the energy consumption and to shorten production times. This idea derives from the generally accepted concept for which the use of recycled materials is environmentally preferable to that of virgin raw materials. This is because the environmental loads associated with the processing of recycled materials are less than those associated with the extraction and processing of virgin raw materials.

As regards glazing manufacturing, EOL CRT glass could be considered as a substitute for non-plastic materials. Moreover, it contains barium, strontium and zirconium oxides, which represent components that are often added to glazes because of their positive effect on glaze quality. Further, the oxides introduced by the CRT glass are very important, e.g. for imparting desired physical and chemical properties to glazes. The relatively high barium oxide content of the CRT glass does not restrict its use either in glass or glaze compositions. In these materials, barium oxide is in fact frequently introduced to enhance some specific property (such as abrasive hardness, modulus of elasticity, refractive index and viscosity, barium glasses are generally soft and long [8]).

4 Glaze Obtainment Process Feasibility

The process feasibility was tested with the contribution of equipment, instrumentation and technological support of a local ceramic glaze producer, within a public/private collaboration [9].

In particular, the suitability of the cleaned EOL CRT TV + PC colour panel glass as a glaze raw material was tested in an industrial, standard glaze for porcelain stoneware and fast single-firing tiles. This glaze contains 35 wt% of glassy frit and other raw materials such as clays and feldspars. 30 wt% of this frit was substituted by panel glass, the identification of this limit percentage is related to the importance of avoiding defects such as bubbling, cracking, warping of the support, which appear for a higher amount. In this way, it has been used to put a base glaze right to be used for the production of pigmented, silk-screened and flame-hardened glazes used industrially for coating floor tiles. The glazes obtained have properties and aesthetic characteristics extremely similar to originals not containing CRT glass; imposing, however, is the particular attention toward the thermal expansion coefficient that tends to be elevated. The modulation of the raw materials in formulating the glaze therefore becomes fundamental to obtain acceptable values of this parameter. Moreover, the presence of small black dots on the surface of the base glaze (probably due to the low efficiency CRT glasses cleaning treatments) has developed the research on rustic or natural stones reproducing glazes. In fact, on these kind of products, the imperfection cited above is masked by the application of other glaze layers, silk-screened and flame-hardened glazes often coloured. Finally, the glazes have been used on single-firing and porcelain stoneware floor tiles and the obtained final products have been characterised by colorimetric analyses and chemical resistance, staining and surface abrasion tests in accordance with ISO testing methods.

From an aesthetic point of view, the products containing CRT glass do not present differences perceptible by eyes with respect to the originals. The results of the chemical resistance tests (UNI EN ISO 10545-13) allowed us to show that there are no appreciable differences between the samples containing CRT glass and standard industrial glazes. The staining tests (UNI EN ISO 10545-14), performed with blue methylene and potassium permanganate ($KMnO_4$), provide an indirect evaluation of the open porosity of the glazed products considered after firing. These tests have shown a high resistance to dirt; the belonging class is 5 (the stains are removed using only water) as the standard one. As regards the surface abrasion tests (UNI EN ISO 10545-7), it was observed that the introduction of CRT glass into the formulation of glazes can improve the resistance to abrasion or confirm the values of the standard product.

This open-loop recycling, successfully experimented, represents the prototyping phase of the project. Present problems associated with the scaling up of this prototype toward real business application are related to the optimising step of the cleaning process. However, the experimental work conducted has been the starting point for the study with the object of providing environmental evaluations. The study was developed following the LCA methodology, using the SimaPro 5.0 software and choosing the Eco-Indicator 99 as an evaluation method. This methodology has already been used in the evaluation of the environmental impact of technologies [10], products [11], recycling processes [12], and to compare different technologies, for example for the production of desktop computer displays [13].

5 LCA Experimental Approach

5.1 Purpose and systems boundaries definition

The study aim is to compare the standard ceramic glaze production, starting from a commercial frit produced inside the industry starting from mineral raw materials, to the formulation of the same glaze in which EOL CRT panel glass is inserted. In this way, the environmental credit has been evaluated between the comparison of the environmental loads of two product systems, one using virgin raw materials and the other using recycled materials.

The system function is represented by 1 kg (also a functional unit) of ceramic glaze produced.

The system boundaries for the glaze production are upstream to the production of the necessary raw materials to produce the glaze, downstream the emissions into the environment related to its production. Otherwise, for the recycling process, the system includes the operations which allow the recycling of the glass, such as the reclaiming, the panel and

funnel separation, the suitable preparation as ceramic raw material (fine and regular grain size) and an important aspect is that the avoided landfilling of the glass is taken into account. Furthermore, the transport of the glass towards the ceramic glaze producer, and the avoided frit production is considered. Hence, the commercial step, the product use and end of life stages of the produced glaze were outside of the system boundary.

The adopted allocation method was a first step in avoiding an allocation through division of the multifunction process (recycling of panel glass) into sub-process (panel glass treatment and glaze production, performed in two different plants, and transport), and separate data collection for each sub-process. This procedure has been adopted because the recycling does not cause a change in the properties of the final material as assessed by the technological characterization. The environmental burdens of each sub-process are allocated between the product outflows from sub-process. In the second step, the environmental burdens of the joint sub-processes are allocated in proportion to the mass of the product.

For the study, the SimaPro 5.0 software is used. Both secondary data (carried out from data base referred to the period 1996–2001 or from previous studies [14,15]) and primary data (supplied by the companies directly or indirectly involved) have been used. The companies directly involved are two Italian companies: one dismantling WEEE plant which has treated about 80,000 PC monitors and 120,000 TV sets in 2001 and a ceramic glaze manufacturer, which produces more than 130 t/month of vitreous products. The companies indirectly involved are located in Italy amount to about 10; they work in different fields related to the different step of the process, such as raw materials extraction and transport, frit production, chemical reagents use, energy producer, water depuration, fume filtration, etc. Weighted impact of the entire product system was assessed using Eco-Indicator 99 as an evaluation method [16]. This method considers three damage categories of potential environmental impacts (each grouping several impact sub-categories together): Human Health, Ecosystem Quality and Resources. The impact assessment is composed of three steps:

– Classification, where data are sorted into classes according to the effect they have on the environment;
– characterization, where the impacts are summed in the framework of the given impact category;
– normalization, where the results of the three damage categories, expressed in three different measurement units, are reported to a reference value;
– evaluation, connected with impact assessment through the introduction of adequate environmental indicators.

The principal modifications brought to the method are:

1. Choice, in the Evaluation phase of the equalitarian prospect, with the attribution of equal weights to the three damage categories (all corresponding to 333,333);
2. consideration of the water consumption of the process in the impact category named Minerals.

5.2 Inventory

In the first part of the study, the attention has been focused on the operation of dismantling, reclamation, cleaning and crushing of the CRT glass by considering the energy consuming area of occupation in the plant, emissions with their eventual treatments, reuse and disposal of the by-product.

In the second part of the study, the industrial glaze production process has been investigated, in particular divided in the following processes:

- Production of glassy frit similar to the CRT panel glass;
- standard glaze production;
- production of a glaze similar for properties to the standard one, but with 30 parts (on 35 total parts of glassy phase) of CRT panel glass as substituted by commercial frits.

For every process, also the sub-processes correlated, such as raw materials extraction, treatment and transport, the emissions in air, water and soil included the treatments of depuration and reuse/disposal of the by-product in those produced, have been taken into account in the study.

For both parts of the study, the LCA involving machineries have not been considered.

Fig. 2 and **3** schematically represent and synthesise the above-mentioned words. All the aspect reported in the figures inside the boundary of the LCA study are therefore included. In

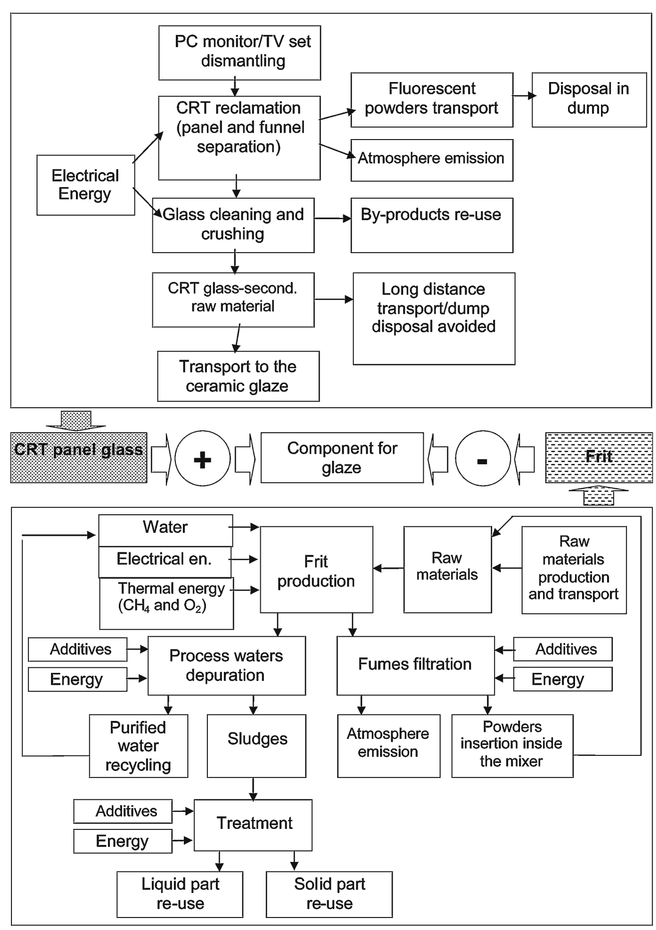

Fig. 2: Experimental recycling (CRT glass as component for glaze) involving the CRT glass preparation or the frit production

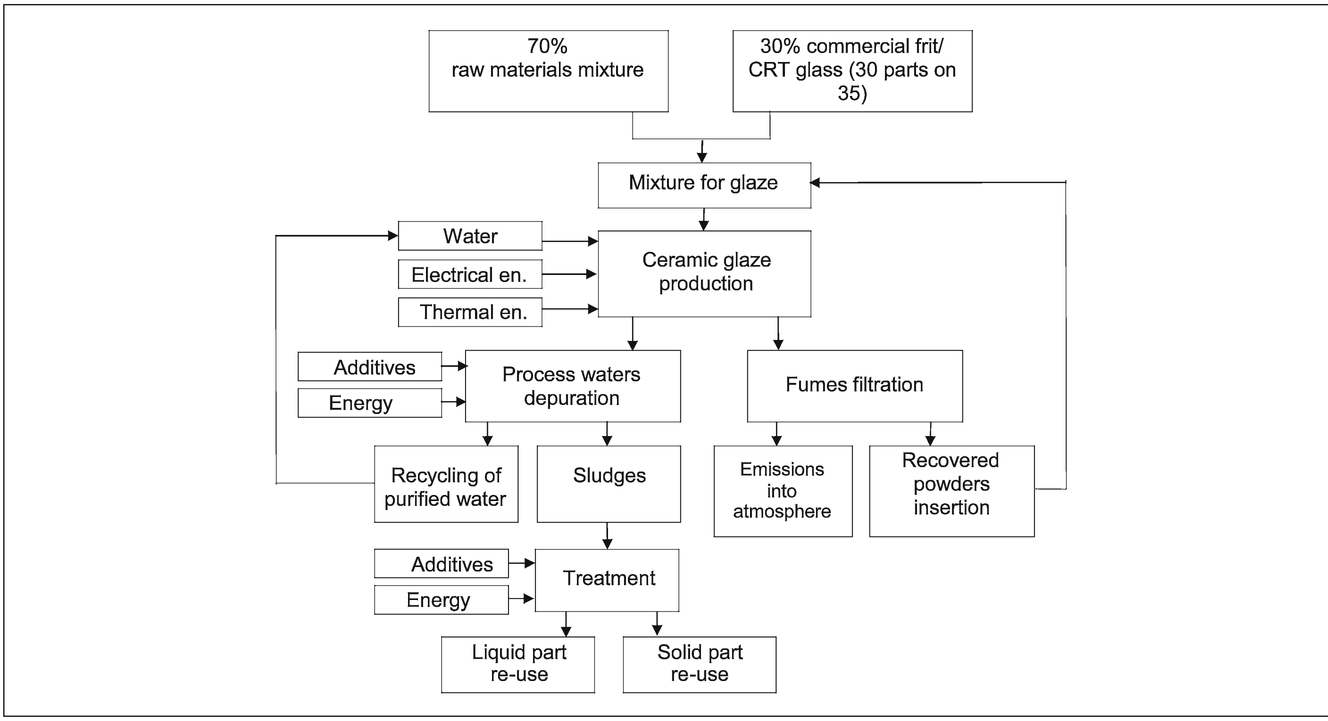

Fig. 3: Flow chart of the ceramic glaze production

particular, Fig. 2 shows the CRT treatment process and the avoided glassy frit production. The average weight of a cathode ray tube is 9.550 Kg and the separation process produces 5,413 kg of panel glass and 2,707 kg of funnel glass.

CRT Reclamation. The reclamation process needs electrical energy of 0.2375 kwh for every CRT treated. The aspirated fluorescent powders are 6.5 g/CRT and they are transported to a dump for dangerous waste by a 28t truck for a distance of 150 km, with a correspondence value of 0.000975 tkm. The emission to air of the reclamation process is 0.05802 g for every CRT.

Glass cleaning and crushing. These processes produced a 95 wt% of glass corresponding to 5.14235 kg of panel glass and 2.57165 kg of funnel glass, while there is a 5 wt% (0.270 kg for every treated CRT) of by-product recycled as ceramic material. These processes together need electrical energy of 0.0375 kwh/kg treated glass. The cleaned panel glass obtained is transported to the ceramic glaze producer which is at a distance of 47.2 km by a 16t truck with a corresponding 0.0472 tkm.

This glass is the secondary raw material for the production of glaze, thus avoiding the production of glassy frit starting from virgin raw material.

The frit production has an output of 82.727% so, for the obtainment of 1 kg of frit, 1.2088 kg of raw materials are necessary. The process consumes both raw materials, such as KNO_3, dolomite, Na + K feldspar, Na feldspar, quartz and borax (for every raw material an LCA of the production and extraction processes is conducted), and thermal and electrical energy. In particular the frit production and bag-filling require an electrical energy of 0.061308804 kwh/kg frit; furthermore, a large amount of thermal energy is needed for the melting of the raw materials. The kilns use O_2 as a comburent for the CH_4, therefore there is a consumption of O_2 (0.497 kg /kg of frit) and of CH_4 (5057.16 kJ/kg of frit). Furthermore, water is consumed in the amount of 0.33908 kg/kg frit and, at the end of the process, a water depuration step is required. This step needs some additives such as NaOH, $AlCl_3$ and polyelettrolites, and 0.01876 kg of sludges are produced for every kg of frit. The sludges are re-used in the production of bricks, while the purified water is reintroduced into the cycle. The fumes are treated with 4.3 x 10^{5-} kg of lime for each kg of the produced frit and filtered with a fabric filter. Since the frit does not contain Pb and F, only emission of particulate is considered. The formed powder is 0.010253 kg for every kg of produced frit, and filters capture 0.00002341 kg of powder. The difference (0.010273 kg) is reintroduced in the cycle as avoided raw materials.

Fig. 3 shows the production of glaze containing panel glass. The CRT glass is introduced as 30 wt% mixed with 70 wt% of other raw materials; therefore, in order to produce 1 kg of glaze, 300 g of panel glass are required, which are obtained from 0.55713 kg of a cathode ray tube with an output of 100%. The water consumption is estimated to be 0.33908 kg/kg glaze. The depuration process with the production of sludges is invariant with respect to the process described above. The energy required is both thermal and electrical, with a consumption of 1608 kJ/kg glaze and 0.1 kwh/kg glaze. This last datum includes both the consumption of production line equipments and the consumption of laboratory, offices and warehouse. This last contribute is 0.006222 kwh. The powder emissions are 0.02171 g/kg glaze and all the consideration above reported for the fume treatments are invariant.

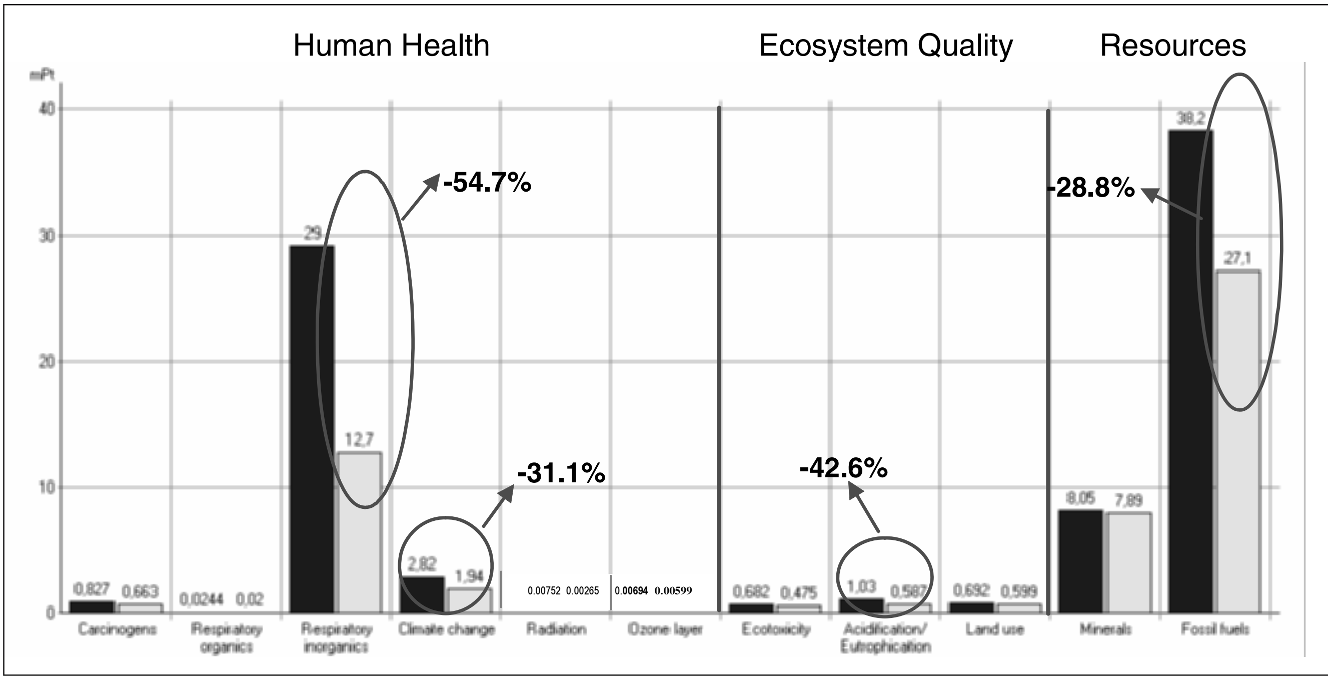

Fig. 4: Evaluation by impact sub-categories related to the production of 1 kg of glaze (black: glaze with commercial frit; grey: glaze with CRT glass)

5.3 Environmental results and considerations

The results of the analysis show that the production of glaze with CRT glass, with respect to standard glaze production, leads to a decrease of the overall potential damage (measured in Points) of 36% and, in particular, a reduction of 53% in 'Human health', 31% in 'Ecosystem quality' and 24% in 'Resources'. This decrease is due to the fact that, notwithstanding the preparation process of CRT glass, it is a complex operation and its transport to the ceramic glaze producer creates an environmental impact, so that the CRT glass utilization positively counterbalances the obtainment process of the frit. This process, in fact, needs both raw materials extraction, which impoverishes the territory, and high energy cost, because of the melting step which consumes high thermal energy in terms of CH_4 and O_2 (5057.16 kJ and 0.497 kg of O_2). On the contrary, the use of a waste material which is already a glass can help firstly to lower this energetic cost and to avoid the extraction operation. The consumption of thermal energy of the glaze productions, in fact, is lower (1,608 kJ) and does not require O_2 contribution. In a more detailed way (**Fig. 4**), the higher environmental impact reduction is in 'Human health' category, where is present the major environmental gain. An improvement is firstly evident in the respiratory, inorganic sub-category (54.7%) related to the lowering (86.5%) of dusts, mainly, and to a lesser amount in NO_x and SO_x. The decrease in dust is due to the avoided production of commercial frit substituted with CRT glass (in particular, due to avoided extraction of raw materials), and NO_x and SO_x are related to the frit production, electricity and thermal energy necessary to melt raw materials to obtain the frit. Further decreases (31.1%) in environmental impact are evident in the climate change sub-category. This is due mainly to the reduction (94.4%) of CO_2 emission correlated with the avoided combustion of the mixture which feeds melting kilns in the frit production. Thus, the damage decrease in 'Ecosystem quality' is prevalently due to the lower of NO_x emissions (93%) by the kilns in the frit production that is evident in the acidification/eutrophication sub-category (42.6%). Finally, the significant saving in 'Resource' category, is principally linked to the fossil fuels subcategory (28.8%), thanks to the CH_4 saving (73.9%) which stokes the melting furnaces.

A remark can be drawn by observing Fig. 4 and it is related to the very low gain in the sub-category minerals with the use of CRT glass; it is explainable by considering the very high impact of the obtainment process of alumina (starting from the extraction of bauxite) on the entire productive cycle. For this reason, being that alumina presents both in the standard glaze and in the CRT glass containing one, no significant variation is observed in this sub-category.

To conclude the environmental impact evaluation of the obtainment of ceramic glazes by CRT glass, it has been performed as a comparison with other commercial products (**Fig. 5**). Fig. 5 shows that environmental impact related to the production of 1 kg of glaze, starting from commercial frit or CRT glass, correspond to those of the production of 0.43 and 0.12 kg of iron, respectively, showing a less environmental impact of the production of CRT containing glaze. The same considerations can be drawn for the other materials listed, confirming again the environmental gain by using EOL CRT glass in the ceramic process. The comparison is made by considering the LCA of these products previously performed with the same criteria of the LCA presented in this work.

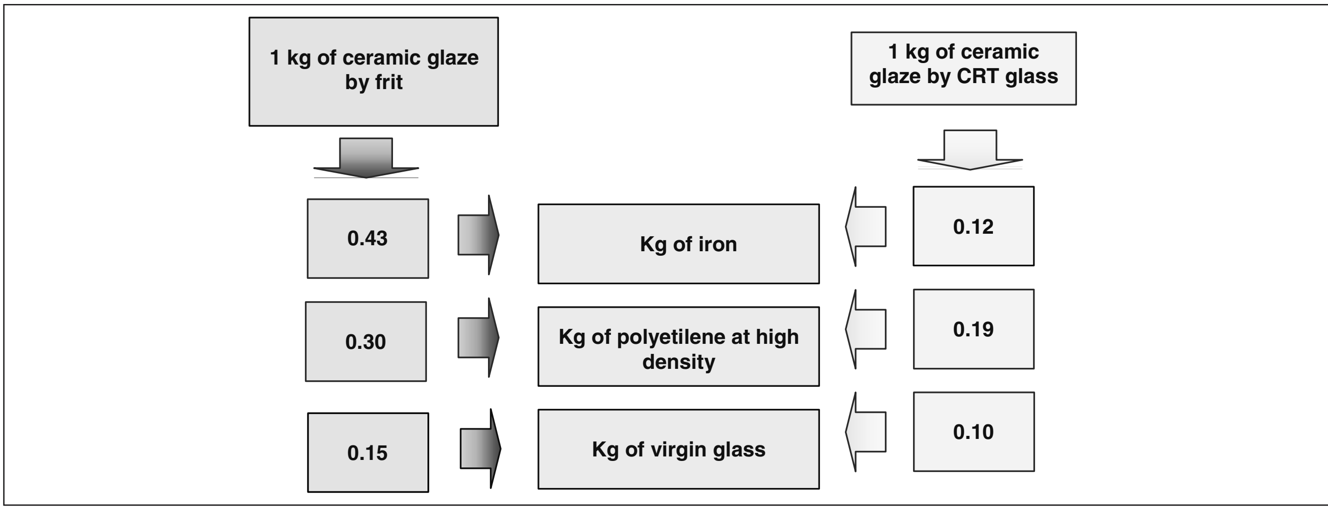

Fig. 5: Comparison of environmental impact value of different products

6 Conclusions and Perspectives

The aim of the study was to support, by an environmental point of view, the open-loop recycling solution in the ceramic field successfully experimented (from a technical point of view) by the authors for the EOL CRT glass. The life cycle assessment approach was used to determine whether the insertion of CRT glass inside a ceramic glaze formulation instead of a commercial frit (made starting from raw materials) would reduce the overall environmental burden of the process. This study has demonstrated that this new open-loop recycling strategy for the CRT glass significantly reduces the environmental impact of the ceramic glaze production process. Another aspect to underline is that CRT glass treatment and the transport of the cleaned glass toward the ceramic glaze producer, in any case, result in having a less environmental impact than the production of the commercial frit.

Furthermore, the decrease of CO_2 emission (94.4%) evident in the climate change sub-category is a very important topic because it is in line with the Kyoto protocol (1997), where significant efforts have been exerted for the reduction of the greenhouse gases emission, notably CO_2. The CO_2 emission is correlated to the combustion of the mixture which feeds melting kilns in the frit production; therefore, the recycling of secondary raw materials, already in a glass state, can reduce the emissions of this gas. This reduction can be termed as environmental credit and it is an example of an allocation of environmental loads in open-loop recycling, where waste from one industrial system are used as raw materials in another product system.

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